

Field induced tricritical behavior in the $S = 1/2$ quasi one-dimensional frustrated Ising antiferromagnet

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The results of extensive histogram cluster heat-bath Monte Carlo simulations on the critical behavior of the quasi-one dimensional Ising antiferromagnet on a stacked triangular lattice are presented. A small applied field is shown to induce a crossover from XY universality to mean-field tricritical behavior. Experimental estimates of critical exponents suggest that these two types of phase transitions are observed in $S = 1$ CsNiCl₃ and $S = 1/2$ CsCoBr₃, respectively. The present results demonstrate that this difference can be explained by an unusual staggered magnetic field arising from quantum exchange mixing previously proposed to account for spin excitations in $S = 1/2$ quasi-one-dimensional Ising antiferromagnets.

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Meaningful comparisons between experimental and theoretical results for critical exponents can be notoriously difficult due to inadequacy of standard techniques to sample thermodynamic quantities close enough to the phase transition region. It is often the case that small perturbations such as weak anisotropy govern the universality of critical phenomena but only very close to the critical temperature (T_N), rendering access through simulations or measurements problematic. Such issues are exacerbated by critical behavior thought to be influenced by magnetic frustration [1]. Additional complications can arise in model systems where exchange interactions are strongly anisotropic or in cases where quantum effects govern spin dynamics. The Ising antiferromagnet on a stacked triangular lattice (ISTAF) with strong c-axis exchange and weak inter-chain exchange interactions is an example of such a model system. Many quasi-one-dimensional (1D) ABX₃ compounds exhibit strong short-range intra-chain magnetic order (SRO) at temperatures well above the onset of true three-dimensional (3D) long range order (LRO) [2]. The present work demonstrates that quantum effects which govern 1D spin dynamics can impact effective critical properties of the 3D system.

Exchange mixing due to an unquenched orbital moment and crystal field effects in quasi-1D $S = 1/2$ ISTAF's has been proposed to give rise to an unusual contribution to the effective Hamiltonian in the form of a staggered magnetic field with a periodicity of two along the c-axis. This term has been employed to explain low temperature neutron scattering spectra in CsCoCl₃ and CsCoBr₃ [3] and Raman data in TiCoCl₃ [4]. Invoking such an effect relies on the well-developed SRO intrinsic to quasi-1D magnetic systems at low T. Its applicability has been questioned, however, due to the fact that the inclusion of such a term in the model Hamiltonian essentially assumes 1D LRO and is thus a mean-field approximation inappropriate for soliton dynamics [5]. The impact of this field term on the measurements of *effective*

critical behavior observed close to T_N is not subject to this restriction and deserves consideration even though it may be strictly irrelevant in the renormalization group sense *at* T_N . In this work, the effect of this quantum staggered field in the $S = 1/2$ quasi-1D ISTAF is mimicked in Monte Carlo (MC) simulations by considering the influence of a small uniform magnetic field applied to a model with ferromagnetic intra-chain interactions with AF inter-chain coupling. No such effective quantum staggered field is predicted to occur in the case of non- $S = 1/2$ STAF's such as ($S = 1$) CsNiCl₃ which has been shown to exhibit integer-spin Haldane-gap phenomena [6].

Critical behavior of the ISTAF have been investigated using numerous theoretical approaches, MC simulations and experimentally. Despite the large number of studies there is still no clear consensus in the literature regarding the critical exponents characterizing the phase transition between the partially disordered phase (PD) where one of the three sublattices remains disordered, and the paramagnetic phase, at T_N . Most modeling results [2, 7] support the notion from symmetry arguments that the transition at T_N belongs to the XY universality class [8]. There have been suggestions, however, from several MC simulations that critical exponents are close to mean-field tricritical [9, 10], but limited statistics and data analysis were used in these investigations. From the experimental standpoint, a number of past neutron scattering studies of CsCoBr₃ and CsCoCl₃ [2] have obtained sets of critical exponents that largely support the idea of XY critical behavior. Intriguingly, recent high resolution neutron scattering experiments on CsCoBr₃ by Mao *et al.* [11] revealed results which suggest with tricritical mean-field behavior and it was speculated that this could be attributed to the anisotropic nature of the exchange interaction or a consequence of the quantum nature of the $S = 1/2$ spins. Experimental data on weakly axial $S = 1$ CsNiCl₃ with a quenched orbital moment [6] are

consistent with XY universality [2, 12]. Both types of experimental systems, strongly Ising and weakly Ising, exhibit magnetic transitions at T_N with the same symmetry and thus should belong to the same universality class.

Motivated by the new experimental data of Mao *et al.*, extensive MC simulations using the efficient Cluster Heat Bath algorithm (CHB)[13, 14], combined with the histogram method[15], were used here to investigate the effects of anisotropic exchange as well as a small applied field on the critical properties of the classical ISTAF. This work serves to extend previous MC simulations of the ISTAF mentioned above, but especially that of Koseki *et al.* [10], where the CHB was used with strongly anisotropic exchange but with no applied field, as well as that of Netz and Berker [16] and Plumer and Mailhot [7] who considered *isotropic* exchange interactions with a nonzero applied field. The present results also complement MC simulations on the frustrated XY-STAF where anisotropy in the exchange interactions was shown to induce a first-order transition [17] in agreement with recent experimental data [18].

MC simulations in the present work used the order-parameter cumulant crossing method[19] to determine the critical temperature of the system. Finite-size scaling analysis at T_N was then used to extract estimates of the critical exponents μ, γ and ν . Both of these techniques have been thoroughly tested on frustrated Ising and Heisenberg spin models [19].

For the purposes of determining the classical critical behavior it is adequate to use a model system with ferromagnetic exchange interactions along the c -axis, J_0 , with or without a small applied uniform field H directed along the c -axis. In addition to near-neighbor AF interactions between chains, J_1 , the effects of a small next-nearest-neighbor exchange coupling, J_2 , was also considered as in previous MC simulations [21]. These effects can be incorporated in the Ising Hamiltonian:

$$\mathcal{H} = - \sum_{\langle i,j \rangle} J_{ij} \sigma_i \sigma_j - H \sum_i \sigma_i \quad (1)$$

where the spin at any site is $\sigma_i = \pm 1$. In zero applied field 4 different cases were investigated with parameters listed in table I. For all cases J_0 and J_2 are ferromagnetic and J_1 is antiferromagnetic. Finite-size scaling analysis was also performed at 5 nonzero values of H for case II. The

TABLE I: Exchange parameters used for the 4 zero-field cases.

Case	J_0	J_1	J_2
I	1.0	-1.0	0
II	10.0	-1.0	0
III	10.0	-1.0	0.1
IV	10.0	-0.3	0.001

isotropic case I ($|J_0| = |J_1|$) was previously investigated using the MC histogram method with periodic boundary

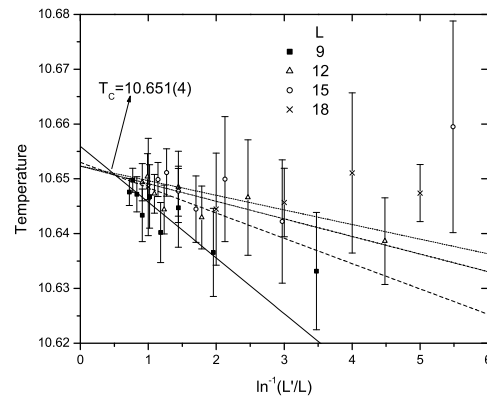


FIG. 1: Results of applying the order-parameter cumulant crossing method to estimate the critical temperature T_N for case II. The straight lines correspond to linear fits to the data with $\ln^{-1}(L'/L) \leq 2.2$.

conditions for an $N = L \times L \times L$ system [7, 20, 21]. Different sets of critical exponents characteristic of different universality classes were obtained in these previous studies, serving to illustrate the sensitivity of analyzing MC simulation data for the frustrated Ising model. Cases II, III, and IV correspond to systems with anisotropic quasi-1D exchange interactions (with $|J_0| \gg |J_1|$) for different values of J_2 . The anisotropic case IV was previously investigated[13] using the CHB algorithm and estimates of the critical temperature and the critical exponents β and ν were obtained using a data collapse method.

The CHB algorithm employs open boundary conditions along the c -axis and periodic boundary conditions in the other directions. For systems with quasi-1D exchange interactions the CHB algorithm is more efficient than the Metropolis algorithm and allows for better statistics when simulating larger lattice sizes. We considered anisotropic lattices with $N = L \times L \times 10L$ and $L = 9, 12, \dots, 33, 36$. These system sizes are smaller than those employed in Ref.[10]. However, a significantly larger number of MC steps were used in any particular run. Averages were also performed over 10 independent simulations using different random initial spin configurations. Estimates of the errors were obtained by taking the standard deviation from the different simulations. Runs of $1 \times 10^5 - 8 \times 10^5$ MC steps were used for equilibration, and $5 \times 10^5 - 1.2 \times 10^6$ MC steps were used to calculate averages of several thermodynamic quantities including the order parameter O , susceptibility χ_1 , specific heat C , energy cumulant U_E , order-parameter cumulant U_M and first logarithmic derivative of the order parameter V_1 , as defined in Ref. [19]. The primary order-parameter O is defined in terms of the \mathbf{Q}_{th} Fourier component of the spin density as $O = |\sum_i \sigma_i \exp(i\mathbf{Q} \cdot \mathbf{R}_i)| / N$ with $\mathbf{Q} = (2\pi/3, 2\pi/3, 0)$ in units of the lattice constants associated with the simple hexagonal structure. In addition, the temperature dependence of the secondary order-parameter O' with the wavevector $\mathbf{Q} = (0, 0, 0)$, corresponding to the uniform magnetization, was also exam-

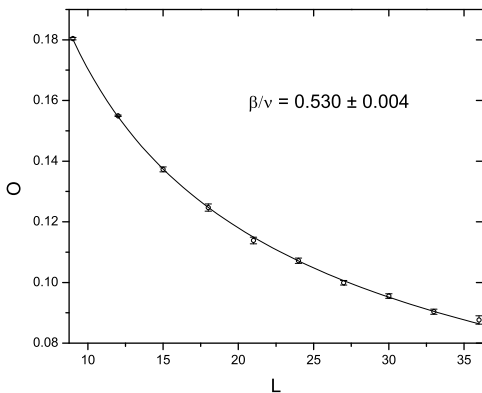


FIG. 2: Finite-size scaling of the order parameter O for case II where error bars on the data represent the standard deviation of the ten runs.

ined. Relevance of this component of the spin density (in zero applied field) on the nature of the critical behavior in these systems has been previously speculated [9, 10].

Accurate estimation of the critical temperature is an essential first step in utilizing the histogram method for determining critical exponents. Temperature sweeps for the $L = 24$ system were initially performed using fewer MC steps to obtain a rough estimate of the transition temperature T_N by locating the maxima of the susceptibility χ_1 and specific heat C . For example, for case IV, temperature scans from $T = 6.8 - 8.0$ in increments of $\Delta T = 0.05$ yielded an estimate of $T_N = 7.40$. To determine the critical temperature more accurately, several histograms were generated at temperatures above and below T_N . In this case, histograms were generated at $T = 7.34, 7.36, 7.38, 7.40, 7.42$ for each lattice size L . The order-parameter cumulant crossing method was used for the sets of parameters of Table I and an illustrative example of the results for case II is presented in Fig. 1. For each case we plot the temperature at which U_M for lattice size L' intersects with the cumulants for $L = 9, 12$ and 15 . Linear fits are made using the results in the asymptotic region (i.e., $\ln^{-1}(L'/L) \leq 2.2$) and an estimate of the critical temperature T_N is obtained from the average value of the crossing points. The error $\pm \Delta T_N$ represents the standard deviation of these values.

The fourth-order energy cumulant evaluated at T_N as a function of system size L was also calculated in each of the four cases. The results extrapolate to $U_E^* = 0.666663(3)$ for $L \rightarrow \infty$ in all cases, as expected for a continuous phase transition. Finite-size scaling analysis at the critical temperature T_N of the various thermodynamic quantities $O \sim L^{-\beta/\nu}$, $\chi \sim L^{\gamma/\nu}$, and $V_1 \sim L^{1/\nu}$, were used to obtain estimates of critical exponent ratios. Representative results are shown in Fig. 2 for the order parameter in case II where the error on the exponent ratio is obtained from the robustness of the fit. Our results show that the secondary order parameter O' is not relevant at T_N for $H = 0$.

In Table II the zero field estimates of the critical tem-

TABLE II: Comparison of the critical temperature and critical exponents determined in this work with other MC studies.

	T_N	β	γ	ν
I	2.926(3)	0.344(7)	1.31(3)	0.671(9)
Bunker et al.[20]	2.920(5)	0.311(4)	1.43(3)	0.685(3)
Plumer et al.[7]	2.9298(10)	0.341(4)	1.31(3)	0.662(9)
Heinonen et al.[9]	2.88	0.19(1)	1.15(5)	
II	10.651(4)	0.355(6)	1.33(3)	0.670(7)
III	11.998(4)	0.358(5)	1.28(3)	0.677(3)
IV	7.406(6)	0.362(7)	1.35(2)	0.673(4)
Koseki et al.[13]	7.34(4)	0.21(1)	1.31(3)	0.70(3)
XY		0.345	1.316	0.671
Tricritical		1/4	1	1/2

perature and critical exponents determined in this work are compared with those obtained by other MC studies. For each case studied, our set of critical exponents indicate 3D XY universality. This is in agreement with symmetry arguments as well as the MC results of Ref. [7] but contrast with the tricritical behaviour seen in Refs. [9] and [13], where less intensive simulations were used.

In order to estimate errors due to the uncertainty in the critical temperature T_N , finite-size scaling analysis was performed at temperatures slightly above and below T_N . In Fig. 3, results are shown for the critical exponents β , γ and ν versus the choice of critical temperature for cases I, II, and IV. We find that the cases with quasi-1D exchange interactions (cases II, and IV) are more sensitive to the choice of critical temperature than the case with isotropic exchange coupling (case I). These results indicate that the critical temperature region is extremely narrow for the quasi-1D models, highlighting the need for care in the analysis and interpretation of both computational and experimental data for such systems.

TABLE III: Critical temperature and critical exponents for several points along the paramagnet phase boundary. The errors on the exponents are estimated from fits performed at $T = T_N \pm \Delta T_N$.

H	T_N	β	γ	ν
0	10.651(4)	0.35(2)	1.33(5)	0.670(7)
0.10	10.651(5)	0.31(3)	1.3(1)	0.63(1)
0.25	10.685(5)	0.28(3)	1.22(6)	0.56(1)
0.50	10.756(5)	0.27(4)	1.02(9)	0.50(2)
1.00	10.891(5)	0.27(7)	1.0(1)	0.48(4)

The effects of a nonzero applied field were examined here for the quasi-1D case II only. This extends the work of Refs. [7, 16] where isotropic exchange was assumed and where it is argued the effect of an applied field is to change the symmetry to that of the 3-state Potts model and hence the transition should be first order within mean field theory. However, a more complicated phase diagram emerged as a result of these earlier MC simulations. The same number of MC steps as in the zero field cases were used in the present study for equilibration and to calculate thermal averages. At field strengths $H = 0.01, 0.25, 0.50$ and 1.0 simulation results for

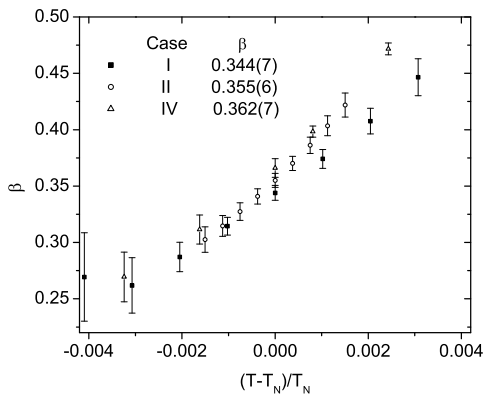


FIG. 3: Values of the critical exponents β for cases I, II, and IV, obtained for different choices of the critical temperature. Results for case III are qualitatively similar and have been omitted for clarity.

the energy cumulant extrapolate to $U_E^* = 0.666663(3)$, suggesting that the phase transitions remain continuous at these lower values. Results for the critical exponents at five field strengths from $H = 0$ to $H = 1$ are shown in Table III. The error for the critical temperature is estimated from the scatter of the crossing points of the order-parameter cumulant data. The errors on the critical exponents are obtained from fits performed at $T = T_N \pm \Delta T_N$. For example, for $H = 0.25$, the fits performed at $T = 10.680$ yielded $\beta = 0.26(1)$, $\gamma = 1.28(4)$, $\nu = 0.56(1)$, whereas those at $T = 10.690$ yielded $\beta = 0.306(6)$, $\gamma = 1.17(2)$, $\nu = 0.57(1)$. These results illustrate the sensitivity of the critical exponents with respect to the choice of critical temperature. The largest source of error on the critical exponents comes from the uncertainty of the critical temperature. The errors from the robustness of the fits are typically smaller. For small values of the applied field ($H = 0.1$) the estimated values of the critical exponents β and ν are slightly lower than the zero field cases. At intermediate field values $H = 0.5$ and $H = 1.0$ the magnetic phase transition is characterized by a set of exponents that are consistent with tricritical mean-field values. Monte Carlo simulations results for $H = 2.0$ reveal a weak first order phase transition where $U_E^* = 0.666642(3)$.

The results of these extensive MC simulations using the CHB algorithm combined with the histogram analysis technique serve to resolve long-standing questions regarding both experimental and previous modeling results on the critical properties of the quasi-1D ISTAF where both XY and tricritical behavior has been reported. This work illustrates the difficulty in extracting reliable estimates of exponents due to the enhanced sensitivity of the critical region when exchange interactions are frustrated and anisotropic. A major focus of the present work has been the careful estimation of errors due to uncertainty in the assumed critical temperature, a feature relevant for the analysis of both modeled and measured data. Our results make clear that the classical model system yields

XY criticality in the case of zero applied field for a variety of assumed exchange interactions, in agreement with most previous MC simulations results and with most experimental results on Ising-like ABX_3 compounds such as $S = 1$ $CsNiCl_3$ [2]. Quantum effects due to SRO inherent in the quasi-1D $S = 1/2$ compounds are mimicked by the addition of a small applied field and are shown to induce a cross-over to tricritical, then first-order behavior. Our results explain recent high-resolution neutron scattering experiments indicating tricritical exponents in $CsCoBr_3$ [11] where the strength of the extra field term had previously been estimated to be $H/J_0 = 0.05$ [3] consistent with our simulations results ($H = 0.5$). This conclusion illustrates that quantum spin effects can control the experimentally accessible effective critical behaviour of quasi-1D ISTAF's. This result is relevant to other systems that exhibit strong SRO before the onset of LRO.

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